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APPROXIMATE DIFFUSION CALCULATIONS FOR THE
FALLING-RATE PHASE OF DRYING

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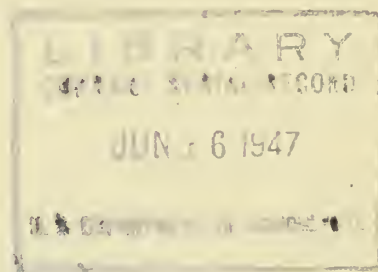
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By W. B. VAN ARSDEL (Active Member)

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ABSTRACT

Some of the observed complexities of drying rate curves have been ascribed to a variability of the diffusivity of the substance with changing moisture content. Quantitative evaluation of the suggestion has been difficult because the generalized diffusion equation is not integrable. Approximate numerical procedures are applied in the present paper to explore the effect of a variable diffusivity in the low-moisture range upon the shape of the rate curve for the infinite slab, the long cylinder, and the sphere. Either vapor pressure or concentration may be used as the driving potential. The effects of changing the thickness of the body and changing the relative importance of the surface mass transfer coefficient are investigated. Typical experimental rate curves, determined during the dehydration of cut vegetables, exhibit curvature of the type predicted by such calculations, and to a degree that might well be expected on the basis of independent measurements of diffusivity in hydrophilic solids.

THE phenomena occurring during the drying of a moist solid, ever since the classical analyses of Lewis, Sherwood, and others, (21)(22)(26)(27)(31) have been divided into two or more distinct phases. The earlier of these, characterized by phenomena similar to those of an evaporating free liquid surface, and known as the constant-rate phase, has presented few difficulties. The one or more later phases of the process, characterized by a steadily decreasing drying rate, exhibits complexities which have not yet been fully unraveled. Various physical mechanisms have been called upon to suggest explanations for the phenomena. Sherwood (31) and Newman (27) employed the diffusion theory first worked out for the unsteady-state conduction of heat, considering moisture concentration gradient as being analogous to temperature gradient. Ceaglske and Hougen (9) and Fisher (13) emphasized the mechanics of flow through capillary pore spaces. Macey (23a) determined the pressure necessary to remove water from moist clays, and identified the diffusional driving force with this pressure. McCready and McCabe (26), and Bateman, Hohf, and Stamm (5) postulated that vapor diffusion occurs within voids and considered that two or more of these mechanisms were effective simultaneously.

During the past five years workers in this laboratory have made a comprehensive investigation of the drying

conditions, such as the use of commercial types of drying trays and heavy tray loadings, which make theoretical analysis of the results difficult. The methods used and the results of the work have been reported by Brown and Kilpatrick (6) and in a series of mimeographed circulars. (36)

Government purchase specifications for dehydrated vegetables required that the moisture content be very low (4 to 6 per cent, depending upon the product). The drying rates in the lower part of the falling-rate phase

FIG. 1. MOISTURE GRADIENTS, CONSTANT DIFFUSIVITY

Slab, half-thickness 4½, $\Delta x = 1$, $s = 1$. $D = 0.20$, k infinite
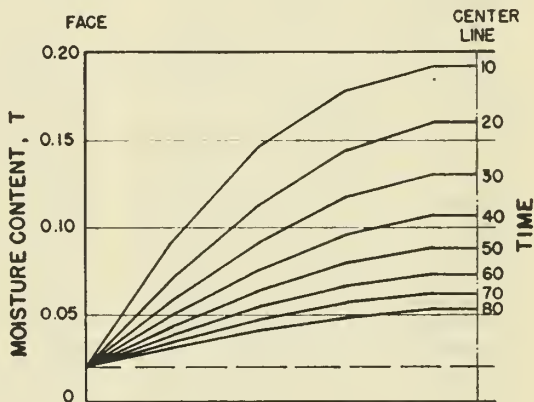
rates of the five or six vegetables which were most important in the wartime dehydration program. The primary purpose of this work was to provide the data needed for rational design of vegetable dehydrators. Consequently most of it was done under

therefore took on great practical significance. Experimental results show, as pointed out by Van Arsdel, (37) that as much time may be required to reduce the moisture content of a vegetable from 7 per cent to 4 per cent as is required to remove the
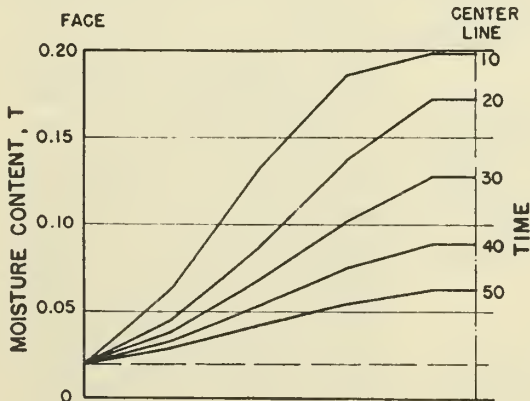

FIG. 2. MOISTURE GRADIENTS, CONSTANT PERMEABILITY

Slab, half-thickness 4½, $\Delta x = 1$, $s = 1$, $P = 4.40 \times 10^{-4}$, k infinite. Vapor pressure isotherm as in Fig. 3
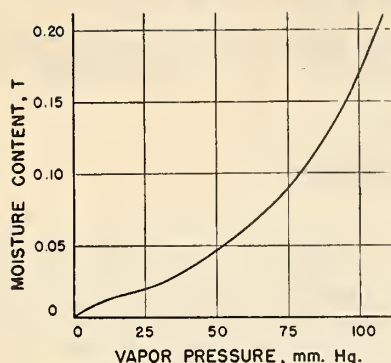
FIG. 3. TYPICAL VAPOR-PRESSURE
ISOTHERM
Dehydrated carrots, 140 Degrees F.

hundred-fold larger amount of moisture from the starting point down to 7 per cent. Since the size of the drier required is directly proportional to the drying time for a specified rate of throughput, the design significance of the final phase is obvious.

During the empirical analyses of experimental drying data in this laboratory it was noted that drying times of vegetable pieces could be correlated satisfactorily by means of nomographs provided the data were regarded as falling into two distinct phases, the dividing line being in the range between 10 and 20 per cent moisture. (6) In the low-moisture phase the characteristics of the data corresponded with those of a system in which the rate of internal diffusion is controlling; that is, neither the air velocity, the type of tray, nor the original depth of tray loading had any further appreciable effect. The data should be amenable therefore to theoretical analysis if the mechanism of moisture transfer in nearly dry materials of this character is similar to any of those postulated by previous investigators. The present study is directed toward the moisture diffusion hypothesis of Sherwood and Newman, and certain modifications of it, rather than the pore flow theory of Ceaglske and Hougen, because the materials in question resemble solid gels in structure.

In this classical diffusion theory the potential producing movement of moisture within the solid piece is identified with a concentration difference. The differential equation expressing the change in concentration at any point with respect to time is as follows for one-dimensional diffusion (the infinite slab):

$$\frac{\partial C}{\partial \theta} = D \frac{\partial^2 C}{\partial x^2} \qquad (1)$$

where D is the diffusivity of moisture in the material. Numerous writers have examined the integrals of this

equation for various important boundary conditions, (3) (7) (15) (16) (21) (27) (31) (32) and the predicted drying rates have been shown to agree reasonably well with experimental data on the drying of some industrial solids. However, when the experimental rate curves are examined in detail, divergence from the theoretical shape is apparent in nearly every case. The divergence is most apparent in the range of very low-moisture contents, where the solutions of Equation (1) predict a direct proportionality between drying rate and free-moisture content. Sherwood, (31) Bateman, Hohf, and Stamm, (5) Hougen, McCauley, and Marshall, (16) and King (20) conclude that diffusivity does not remain constant, but is a function of moisture content. The mathematical complexity of the resulting case is such that no very useful analytical solutions for the diffusion equation have been found, and the observation has remained a purely qualitative one.

There is now ample evidence that the diffusivity of moisture in hydrophilic substances indeed is highly dependent upon the moisture content. The results of Doty, Aiken, and Mark (10) indicate that the diffusivity of moisture through regenerated cellulose changes by at least forty-fold within a comparatively narrow range of moisture contents. King, (20) studying the moisture relations of keratin proteins, such as horn, found more than a hundred-fold change. The failure of experimental drying curves to follow the course predicted by the simple diffusion theory is scarcely surprising.

Approximate numerical solution of the diffusion equation offers no great difficulty, and it possesses the advantage that some of the simplifying assumptions that are necessary to the analytical integration of Equation (1) may be dropped if desired. The two most important of these assumptions, from our viewpoint, are the following:

1. The potential producing diffusion is a concentration difference.
2. The diffusivity is independent of the moisture content.

Some other assumptions that must be introduced into the analytical treatment become unnecessary in the numerical method. Such is the case, for example, with respect to the shrinkage in dimensions as moisture is lost, to the existence of temperature gradients within the piece, and to nonhomogeneity of the material and variation of diffusivity with direction in the piece. The limiting factor, in most practical cases, is likely to be the difficulty of experimental

determination of the necessary data. These further complexities are not considered in the present study. Shrinkage and the existence of temperature gradients may be neglected in fact without appreciable error if attention is directed solely to the region of low-moisture content.

The Driving Force for Diffusion

Equation (1) is derived from an expression analogous to Newton's law of cooling and to Ohm's law. The common characteristic of these two laws is that they equate a rate of flow to a certain potential difference divided by a resistance (or multiplied by a conductance). The potential difference is pictured as an unbalanced force, producing motion against something comparable to friction.

Several characteristics of moist bodies, besides concentration differences, might be pictured in the role of diffusion-producing potentials. An obvious one is the activity of the moisture at any given point, as measured by its equilibrium vapor pressure there. The Ohm's law type of expression still can be used, provided the conductance term is written also in appropriate new units. It is now customary in diffusion studies to name this conductance variable the *permeability* when the potential is vapor pressure, while *diffusivity* is the corresponding term when the potential is concentration. Then (*see* McCready and McCabe (26)) the general equation (written to allow for a variable permeability) will become:

$$\frac{\partial C}{\partial \theta} = \frac{\partial}{\partial x}\left(P \frac{\partial p}{\partial x} \right). \quad (2)$$

Or, in the notation more commonly used in drying studies,

$$\frac{\partial T}{\partial \theta} = \frac{1}{s} \cdot \frac{\partial}{\partial x}\left(P \frac{\partial p}{\partial x} \right). \quad (3)$$

It must be emphasized that the choice of potentials and conductances is, in the absence of a temperature gradient, only a matter of convenience; if the proper units are consistently used the solutions of the basic equation are identical. This is not to say, however, that one choice may not be better than another theoretically. It is to be expected that some one of the hypothetical mechanisms will be the most nearly in accord with other physical knowledge about the substance. Three different lines of investigation may be pertinent to this theoretical question.

Consider first the temperature coefficient of diffusion rate. If concentration is considered as the potential then the entire effect of a change in

temperature of the system must be ascribed to a change in the diffusivity of the solid. If vapor pressure is the potential, on the other hand, some of the change in rate of transfer must be ascribed to a change in the potential difference and much less to a change in the properties of the solid. Measurements of the effect of temperature on swelling, molecular spacing, or other properties may support one of these conclusions against the other.

Or again, consider the variability of diffusivity (and permeability) with moisture content. In general, the water vapor pressure of hydrophilic substances does not obey Henry's law. The two constants, D and P, are related to each other through the actual vapor-pressure isotherm of the substance,[*] and in all the common hydrophilic substances that isotherm is strongly curved, generally sigmoid in shape. The consequence of this curvature is that if either D or P is independent of moisture content, the other must vary with moisture content; or, more generally, the variations of D and P with moisture content will follow quite different courses. Once again, measurements of other physical properties of the substance may accord better with one of these pictures than with the other.

Diffusion in nonisothermal solids has received only the most superficial study. The few observations [1] [3] [23] are only semiquantitative, and the conclusions drawn are, in part, contradictory. It does appear to be certain, however, that moisture transfer occurs within a uniformly moist body under the action of an imposed temperature gradient. In this case, surely, concentration difference may not be regarded as the potential term, for there is no concentration difference. Apparently, the converse case never has been investigated; consider, that is, whether steady flow of moisture through a diffusion barrier would occur if a temperature gradient were maintained in the barrier but the vapor pressure of moisture were maintained the same on both faces of the sheet. In the latter case there would be no vapor-pressure gradient,

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[*] If the units used are consistent, the constants are related as follows:

$$D = \frac{P}{s} \cdot \frac{dp}{dT} \qquad (3a)$$

A consistent set of units would be, for example, for D: sq.cm. per sec.; for s: g. per cu.cm.; for p: mm. Hg.; for T: dimensionless; for P: g. per sec. through 1 sq.cm. under a gradient of 1 mm. Hg. per cm. thickness (or (g.)/(sec.) (sq.cm.) (mm.Hg./cm.)). Since the slope, dp/dT, of the vapor-pressure isotherm changes with moisture content, the relation between D and P is likewise variable.

FIG. 4. DRYING RATES, CONSTANT DIFFUSIVITY AND CONSTANT PERMEABILITY

Slab, half-thickness 4½, $\Delta x = 1$, $s = 1$. Curve A, $D = 0.20$, k infinite. Curve B, $P = 4.40 \times 10^{-4}$, k infinite, vapor pressure as in Fig. 3
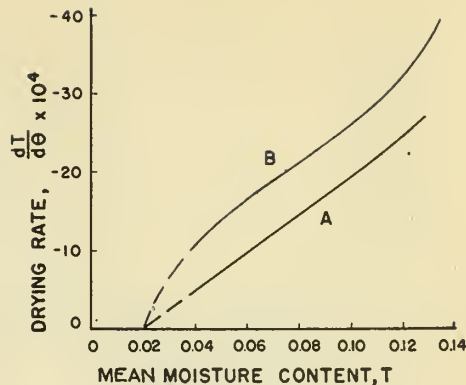
but there would be in general, a concentration gradient across the sheet. Certain of the lines of investigation referred to in this and the preceding paragraphs are being pursued by other workers in this laboratory.

If the mechanism of diffusion is pictured in the manner suggested by Barrer [3] [4] and Stearn and

FIG. 5. MOISTURE CONTENT AT CENTER-LINE

Conditions as in Fig. 4. Curve A, diffusivity constant. Curve B, permeability constant

Eyring [33] and further developed by Cassie [8] and King [20], the form of the vapor-pressure isotherm should be correlated with that of the permeability vapor-pressure relation. This approach, which is that of statistical mechanics, postulates that water molecules which have absorbed more than a certain minimum activation energy are able to move across energy barriers characteristic of the molecular structure of the solid. Application of the Clausius-Clapeyron relation to the vapor-pressure isotherms of various organic materials shows, however, that the binding energy for water increases substantially and rapidly at low-moisture contents; the calculated ratio of the latent heat of vaporization of water from dehydrated carrots to the latent heat of vaporization of free water at the same temperature rises from very nearly

unity at 8 per cent moisture to about 1.60 at 2 per cent moisture. The height of the postulated energy barriers evidently should be substantially greater at the lower-moisture level, and the permeability therefore would be lower.

Materials, such as dehydrated vegetables, are highly complex, multicomponent systems. Their diffusion characteristics must be expected to be a composite of the characteristics of crystalline or glassy sugars, gelled starch, coagulated proteins, cellulosic cell walls, pectins, lipids, and other constituents. Even if each of these constituents should exhibit a relatively sharp break in properties at some level of moisture content, the combined effect of all of them would be to produce a more gradual change. To the extent that the individual constituents are segregated, rather than in solid solution in one another, various levels of moisture content may exist side by side in equilibrium because of the different activity of moisture sorbed in the different constituents.

Approximate Methods of Integration

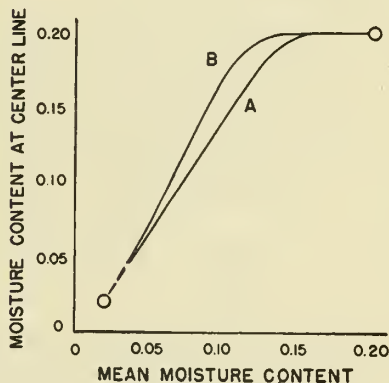Approximate methods for the solution of the diffusion equation have recently been the subject of publications
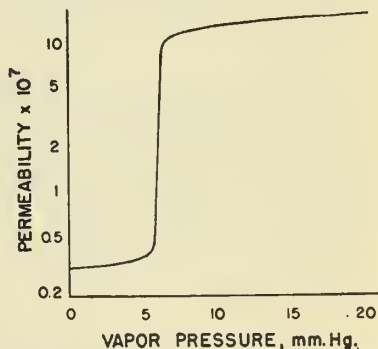

FIG. 6. WATER VAPOR PERMEABILITY OF CELLOPHANE, 25 DEGREES C.

Deduced from data of Doty, Aiken, and Mark, [10] Permeability in cc. water vapor N.T.P./sec. through 1 cm.² for gradient 1 cm. Hg./mm. thickness
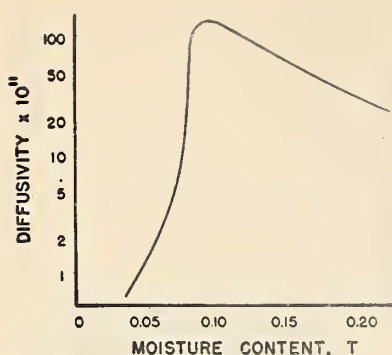
FIG. 7. MOISTURE DIFFUSIVITY IN CELLOPHANE, 25 DEGREES C.
Deduced from. (10) and (17) Diffusivity in cm.²/sec.

by Dusinberre, (11) Paschkis and Baker, (29) and others. (12) (32) The graphical method described by Schmidt (30) was shown by Dusinberre to constitute a special case, and to possess decided limitations. The modification of Schmidt's method to take account of a variable diffusivity, referred to briefly by Sherwood and Reed, (32) appears from unpublished studies made by A. H. Brown, of this laboratory, to be of doubtful general validity, and at any rate to be inapplicable if the diffusivity varies substantially. A semigraphical method proposed by Dusinberre to handle the variable diffusivity case likewise has been found unsatisfactory if the diffusivity varies widely and abruptly.

Numerical methods of solution are based upon mass balances applied to small, but finite, elements of the solid for small, but finite, increments of time. As in Dusinberre's analysis, we make the following assumptions:

1. A time interval can be chosen sufficiently small that there is a negligible error in using the initial potential gradient to compute the flow of moisture during this interval. (Note that even though the potentials may be changing rapidly in two adjoining elements, the difference between the two potentials, which determines the gradient, changes very much more slowly.)

2. The time interval is sufficiently small that there is negligible error in neglecting the effect, during that interval, of any elements beyond the immediately adjacent ones.

3. The dimensions of the volume element are sufficiently small that there is negligible error in using the potential at its center point to compute the moisture content of the element, or vice versa.

Following Dusinberre's suggestion, we find it convenient to make the outermost volume elements only half as thick as the interior ones, so that the "center points" of these outermost elements will fall on the surface of the body. Film coefficients of mass transfer to the air phase are then readily introduced into the calculation.

The Working Equations

Consider one-dimensional diffusion in an infinite slab. Then, using vapor pressure as a typical potential term, the change in moisture content, ΔT, in the nth element from the surface during the time interval $\Delta \theta$ will be:

$$\Delta T_n = [P_{n+1,n}(p_{n+1} - p_n) + P_{n-1,n}(p_{n-1} - p_n)]\frac{\Delta \theta}{s(\Delta x)^2}. \quad (4)$$

Since the permeability may undergo a radical change within the range of p_{n-1} to p_{n+1}, the proper evaluation of the terms within the bracket is crucial. In general,

$$P_{m,n}(p_m - p_n) = \int_{p_n}^{p_m} P\,dp. \quad (5)$$

The permeability will be determined usually as an empirical function of moisture content, but equally well may be plotted or tabulated as a function of vapor pressure by use of the known moisture content-vapor pressure relationship. If Equation (5) is put into the form,

$$\int_{p_n}^{p_m} P\,dp = \int_{p_o}^{p_m} P\,dp - \int_{p_o}^{p_n} P\,dp, \quad (6)$$

where p_o is any convenient base value of the vapor pressure, the value of the graphically or numerically determined integral may be plotted against vapor pressure and the terms in Equation (4) then may be computed readily.

The change in moisture content in the surface element may be similarly shown to be:

$$\Delta T_1 = 2[P_{21}(p_2 - p_1) + k\Delta x(p_a - p_1)]\frac{\Delta \theta}{s(\Delta x)^2} \quad (7)$$

Thus, if the vapor pressures at the reference points throughout the slab at a given instant of time are known, the change of moisture content at each of these points in a time interval $\Delta \theta$ can be calculated. Two different procedures may be used for computing successive states of the system: (a) The changes in T may be followed from step to step, and when the new set of values of T has been established, corresponding values of p may be read from the vapor-pressure isotherm; or (b) only the changes in p may be followed from step to step; a curve giving dp/dT may be derived from the isotherm; then:

$$\Delta p = \frac{dp}{dT}\Delta T. \quad (8)$$

The second procedure is generally preferable. Values of T throughout the slab do not need to be evaluated at every step, but can be read from the isotherm at the time intervals for which they are wanted.

Equation (8) is relatively precise for small values of ΔT, but a second-order correction may be applied without difficulty if desired; instead of reading dp/dT for the value of p at the beginning of the time interval, it can be read at an estimated mean value of p for the interval, calculated mentally by subtracting one-half of the value of Δp found for the preceding time interval.

If an unduly large value of $\Delta \theta$ is used in the stepwise calculation, or an unduly small value of Δx, the calculations will exhibit an instability which eventually leads to absurd r‐ -

TABLE 1.

Mean moisture content of infinite slab. Initial moisture content uniform, $T = 0.2000$. Equilibrium moisture content, $T_e = 0.0200$. Flow proportional to concentration gradient. Diffusivity constant, $D = 0.20$. Surface film coefficient, $k = \infty$. $s = 1$, $\Delta x = 1$. Half-thickness of slab divided for approximate calculation into five elements.

Time	15	30	50	60	70	80	85
Approximate	0.1221	0.0904	0.0632	0.0538	0.0464	0.0406	0.0382
Exact	0.1216	0.0903	0.0632	0.0538	0.0465	0.0407	0.0384
Error	+.0005	+.0001	0	0	—.0001	—.0001	—.0002

TABLE 2.

Mean moisture content of infinite slab. Initial moisture content uniform, $T = 0.2000$. Equilibrium moisture content, $T_e = 0.0200$. Flow proportional to concentration gradient. Diffusivity constant, $D = 0.20$. Surface film coefficient, $k = 0.10$. $s = 1$, $\Delta x = 1$. Half-thickness of slab divided for approximate calculation into 5 elements.

Time	30	50	75	100	120	140	160	180
App.	0.1387	0.1127	0.0881	0.0700	0.0590	0.0504	0.0438	0.0385
Exact	0.1402	0.1142	0.0891	0.0710	0.0597	0.0510	0.0440	0.0388
Error	—.0015	—.0015	—.0010	—.0010	—.0007	—.0006	—.0002	—.0003

sults. It is as though a resonant oscillation, with rapidly increasing amplitude, were set up in the system. Dusinberre (11) proposes the use of a modulus which will assure a proper choice of time and space increments. In general, the time increment can be increased gradually as the system approaches equilibrium conditions. The resulting saving in labor makes it worth while to try this expedient at intervals. If the oscillation effect then begins to appear, the computer can return to the shorter time interval and the divergence will be damped out quickly.

Lederer (21) and Newman (27) have deduced a theorem which, at least in the classical diffusion theory, makes it possible to pass readily from a solution for a one-dimensional system (infinite slab) to solutions for corresponding two-dimensional (infinitely long rectangular strip) and three-dimensional (rectangular block) systems. Unfortunately, the demonstration appears to be valid only for a constant diffusivity. The approximate numerical method becomes excessively laborious when applied to the strip and the cube. Working equations for these cases are reproduced in the Appendix to this paper. On the other hand, calculations for the long cylinder and the sphere are little more difficult than the one-dimensional case. The finite volume elements may be taken as concentric cylindrical or spherical shells, respectively. Mass balances then lead to the following general expressions:

For the long cylinder:

$$\Delta T_n = \frac{2}{2a+1}\left[aS_n^{n+1} + (a+1)S_n^{n-1}\right]\frac{\Delta\theta}{s(\Delta r)^2},$$
(9)

where $a = \frac{r_n}{\Delta r}$, r_n being the inside radius of element n, and the symbol S_n^m signifies the integral:

$$S_n^m = \int_{p_n}^{p_m} P\,dp$$

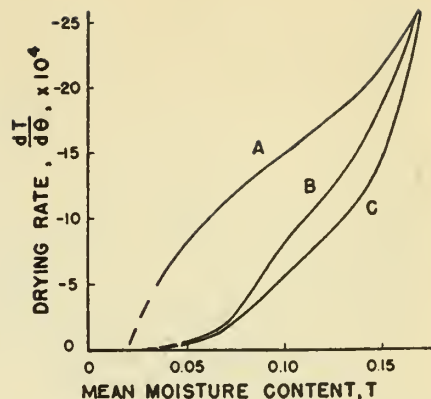For the special case of the central core, where $a = 0$, Equation (9) reduces to:

$$\Delta T_m = 2S_m^{m-1} \cdot \frac{\Delta\theta}{s(\Delta r)^2}$$
(10)

and for the outside shell, assumed to have only half the thickness of the others,

$$\Delta T_1 = \frac{2}{a+\frac{1}{4}}\left[aS_1^2 + k(\Delta r)(a+\frac{1}{2})(p_a-p_1)\right]\frac{\Delta\theta}{s(\Delta r)^2}$$
(11)

For example, if the cylinder is divided into five concentric elements, the outermost having only half of the standard thickness, Δr, the working equations are as follows:

$$\Delta T_1 = [1.882 S_1^2 + 2.118\,k(\Delta r)(p_a - p_1)]\frac{\Delta\theta}{s(\Delta r)^2}$$

$$\Delta T_2 = (0.858\,S_2^3 + 1.142\,S_2^1)\frac{\Delta\theta}{s(\Delta r)^2}$$

$$\Delta T_3 = (0.800\,S_3^4 + 1.200\,S_3^2)\frac{\Delta\theta}{s(\Delta r)^2}$$

$$\Delta T_4 = (0.667\,S_4^5 + 1.333\,S_4^3)\frac{\Delta\theta}{s(\Delta r)^2}$$

$$\Delta T_5 = 2(S_5^4)\frac{\Delta\theta}{s(\Delta r)^2}$$

The mean moisture content of this cylinder may be computed as follows:

$$T = \frac{4}{81}(4.25\,T_1 + 7T_2 + 5T_3 + 3T_4 + T_5)$$

For the general element of the sphere:

$$\Delta T_n = \frac{a^2 S_n^{n+1} + (a+1)^2 S_n^{n-1}}{a^2 + a + \frac{1}{3}} \cdot \frac{\Delta\theta}{s(\Delta r)^2}$$
(12)

For the central element:

$$\Delta T_m = 3S_m^{m-1} \cdot \frac{\Delta\theta}{s(\Delta r)^2}$$
(13)

For the surface element (half-thickness):

$$\Delta T_1 = 2\left[\frac{a^2 S_1^2 + k(\Delta r)(a+\frac{1}{2})^2(p_a-p_1)}{a^2 + a/2 + 1/12}\right]\frac{\Delta\theta}{s(\Delta r)^2}$$
(14)

The working equations for a sphere divided into five concentric elements are as follows:

$$\Delta T_1 = [1.770\,S_1^2 + 2.240\,k(\Delta r)(p_a - p_1)]\frac{\Delta\theta}{s(\Delta r)^2}$$

$$\Delta T_2 = (0.730\,S_2^3 + 1.300\,S_2^1)\frac{\Delta\theta}{s(\Delta r)^2}$$

$$\Delta T_3 = (0.632\,S_3^4 + 1.420\,S_3^2)\frac{\Delta\theta}{s(\Delta r)^2}$$

$$\Delta T_4 = (0.429\,S_4^5 + 1.714\,S_4^3)\frac{\Delta\theta}{s(\Delta r)^2}$$

$$\Delta T_5 = 3 S_5^4 \cdot \frac{\Delta\theta}{s(\Delta r)^2}$$

$$T = \frac{1}{91.2}(27.2T_1 + 37T_2 + 19T_3 + 7T_4 + T_5).$$

Precision of Approximate Method

Partial tests of the precision of the numerical method of computation have been made by comparison with the results of analytical solution in certain simple cases. Tables 1 and 2 show that the accuracy of estimation of mean moisture content is amply good for most purposes.

Experience with the routine computation of a considerable number of cases indicates that an experienced calculating machine operator can complete about fifty steps per day in a problem involving only one-dimensional diffusion, the solid body being divided into five-volume elements, and the permeability and vapor-pressure functions being given as empirically plotted curves. Simple computations such as those leading to Tables 1 and 2 proceed considerably faster.

Effect of Diffusion Variables on Drying Rate

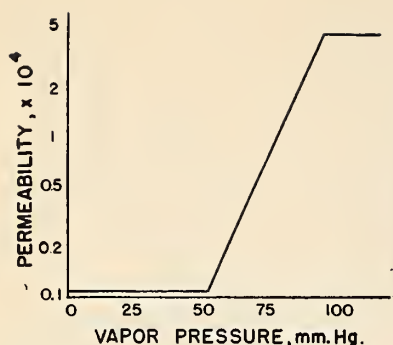It may not be apparent immediately that the drying phenomena that

FIG. 9. FIRST ARBITRARILY ASSUMED
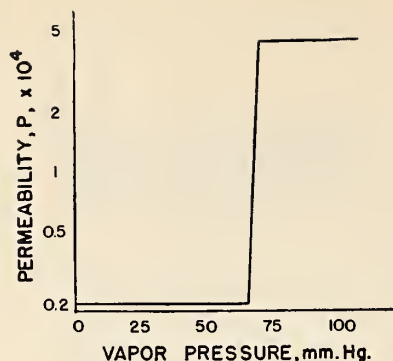PERMEABILITY

FIG. 10. SECOND ARBITRARILY ASSUMED
PERMEABILITY

would be described by Equation (1) are not identical with those corresponding with Equation (2) if the diffusivity be taken as constant in the one case and the permeability as constant in the other. Figures 1 and 2 show successive moisture gradients through the half-section of an infinite slab for the two cases. Figure 3 shows the vapor-pressure isotherm that was used in calculating the gradients for Figure 2; it is that of one sample of dehydrated carrots at 60° C. Figure 4 shows the relation of drying rate to mean moisture content for the two cases, and Figure 5 the relation between mean moisture content and the moisture content at the center of the slab. The sigmoid shape of some of the gradients in Figure 2 is striking. Gradients having this shape have been found experimentally by several workers (5) (9) (16) (19) (26) (31) (34) but Ceaglske and Hougen believed the phenomenon to be "inconsistent with the behavior of diffusion in slabs (9)." Evidently it is consistent, however, with a diffusion equation that is not too rigidly restricted by simplifying assumptions.

*Variation of Diffusivity
and Permeability*

The measurements of moisture transfer through cellophane reported by Doty, Aiken, and Mark (10) do not lend themselves to precise calculation of permeability as a function of moisture content because the humidity gradients used were so great that a wide range of moisture contents must have been present within a single sheet. The relation shown in Figure 6 may be deduced, however, as at least consistent with the data. Figure 7 shows the corresponding relation between diffusivity and moisture content, calculated from Figure 6 and a vapor-pressure isotherm for viscose rayon reproduced in International Critical Tables.(17) The statement made earlier that the choice of potential and conductance terms is

mainly a matter of convenience may be illustrated from these data. All solutions of the following form of the diffusion equation:

$$\frac{\partial T}{\partial \theta} = \frac{\partial}{\partial x}\left(D \frac{\partial T}{\partial x} \right), \quad (15)$$

will be identical with the solutions of Equation (3), namely:

$$\frac{\partial T}{\partial \theta} = \frac{1}{s} \cdot \frac{\partial}{\partial x}\left(P \frac{\partial p}{\partial x} \right),$$

if the values of D from Figure 7 are used in the former and the values of P from Figure 6 in the latter.

The effects of the following variables on drying rate in the lower part of the falling-rate phase are studied readily by the use of the approximate numerical procedure.

Change of permeability with moisture content.

Form of the vapor-pressure isotherm.

Change of the ratio between surface film coefficient and permeability.

Change in the shape of the body (practically limited to the slab, the cylinder, and the sphere).

Change in the thickness of the body.

Figures 1, 2, 4, and 5, already discussed, show the variations that may be expected to follow from the use of different characteristics for the diffusion potential term if an unvarying conductance term be assumed.

*Change of Permeability with
Moisture Content*

Figure 8 compares computed drying rates for the infinite slab, assuming three quite different permeability functions. All are based on vapor pressures as the potential term, and the isotherm is that reproduced in Figure 3. For curve A the permeability was assumed to remain unvarying with moisture content, while for curves B and C the functions represented in Figures 9 and 10, respectively, were assumed. The order of

magnitude of the changes was suggested by Doty, Aiken, and Mark's results for cellophane, but the functions were given simple geometrical shapes so that the relation between the shape of these curves and that of the derived rate curves could be easily analyzed.

Figure 11 shows a group of moisture gradients in the slab for the conditions of curve B in Figure 8. Gradients like that at time 100 have been observed experimentally by many investigators. The material is "case-hardened."

The peculiar shapes of the rate curves indicate how profoundly the drying rate may be affected by changes in permeability that are, in general, known to occur. A note of caution on the interpretation of experimental drying rate curves is strongly suggested. The existence of a substantially straight segment of the falling-rate curve is no guarantee that the straight line may be extrapolated all the way to the base line. Again, since an estimation of drying time almost invariably is the ultimate object, any line of best fit which is to be drawn through a set of points representing successive experimental drying rates should deviate from the points by a smaller and smaller average distance as zero rate is approached.

Rate curves B and C in Figure 8 have no simple relationship to the permeability functions on which they were based. This is, of course, only what might have been expected; at any given moment the drying rate is the result of a complex internal situation which finds some parts of the solid still in the high-permeability range while others have already dried to a condition of low permeability. The effect of the sharp break in permeability shown in Figure 9 at a vapor pressure of about 70 mm. (corresponding to a moisture content $T = 0.0800$) is apparent in the rate curve at a mean moisture content far above $T = 0.0800$. No presently known procedure can be used to carry the calculation in the opposite direction—that is, to deduce quantitatively a permeability function from a given drying rate curve.

*Form of the Vapor-Pressure
Isotherm*

Figure 12 compares the computed drying rates for an infinite slab, using three different vapor-pressure isotherms, the permeability function being assumed as in Figure 9 in all three cases. The three isotherms, which are drawn in Figure 13, were taken from experimental data on three different samples of dehydrated

vegetables; the temperature is assumed in all cases to be 140 degrees F.

Effect of Surface Film Coefficient

Figures 14 and 15 show drying rates for an infinite slab as affected by changes in the ratio between the surface film coefficient and the permeability. In Figure 14 the permeability is assumed to remain unchanged at all moisture contents, while in Figure 15 it is assumed to change twenty-fold within a narrow range, as in Figure 9. By comparison of the two figures it may be seen that when the permeability is variable it is the ratio of the surface coefficient to the low range of permeability values that is controlling, even in the higher levels of moisture content.

Drying Rates of the Slab, Cylinder, and Sphere

Figure 16 compares drying rates for an infinite slab, an infinitely long cylinder, and a sphere, all of the same half-thickness. The vapor-pressure isotherm of Figure 3 and the permeability function of Figure 9 were used in all three cases.

The classical diffusion theory, assuming a constant diffusivity, predicts that in the lower, essentially linear, portions of the rate curves the ratio of the drying rates for three such bodies will be approximately 1:2.3:4.4. The introduction of a variable permeability destroys the linear character of the curves and leads to somewhat erratic values of these ratios, depending to some extent upon the form of the permeability and vapor-pressure functions. In this example the ratios in the lower portions of the curves are approximately 1:2.7:4.9.
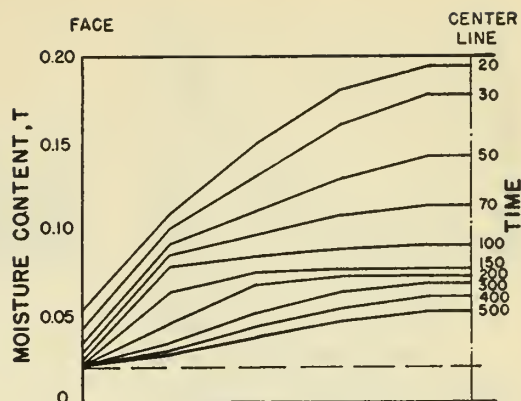
Effect of Thickness

Classical drying theory predicts that if the surface film resistance is negligible the drying rate will vary inversely as the square of the thickness. Figure 17 compares the drying rates of two slabs whose thicknesses are in the ratio of $4\frac{1}{2}:9\frac{1}{2}$; the surface film resistance is negligible (coefficient infinite) and the permeability remains constant at all moisture levels (the diffusivity therefore changes markedly with moisture content). Comparison of the ordinates of the two curves shows a constant ratio of 4:46, which is, within the limit of error, the square of the ratio of thicknesses. The classical rule is therefore valid for the more general case. This conclusion may be demonstrated also analytically.

If the drying rate that is being measured is that of an assemblage of pieces which vary somewhat in thickness (as is always the case in the drying of prepared vegetables) the resulting rate curve is, of course, a composite. The effect will be to mask such sharp changes in curvature as appear in curve *B* of Figure 8. For example, suppose 50 per cent by weight of a mixture consists of pieces which alone would have the drying rate of curve *B*, 30 per cent consists of pieces having only half the standard thickness, and 20 per cent consists of pieces having only one-quarter the standard thickness. Figure 18 shows how the composite drying rate compares with the standard drying rate, Curve *B*. (This calculation ignores the mutual effects of different pieces, and therefore can only hold for a very lightly loaded tray.)

Shapes of Typical Experimental Curves

Figure 19 shows several experimental rate curves, taken at random from the large number determined during the past few years in this laboratory. In these experiments the materials were dried in shallow layers on screen-bottom trays, using cross-flow of air. The results have more general significance, however, because in this low-moisture region the man-

ner of exposure to the air stream has little or no effect on the drying rate. The literature of drying theory provides few data suitable for comparison. Some of the drying experiments (on riced white potatoes), reported by Marshall (24) were carried to a moisture content below 10 per cent, and the few points in this low-moisture region indicate curvature of the rate curves generally similar to the curvature appearing in Figure 19.

The shapes of these curves are strongly reminiscent of some of the theoretical curves — for example, curve *C* in Figure 8. Variation of permeability no greater than has been demonstrated already for cellophane and horn membranes would account without difficulty for the experimental rate curves.

Conclusions

Approximate numerical solutions of the general diffusion equation for the infinite slab, the long cylinder, and the sphere have a useful degree of precision without being excessively laborious.

Reasonable assumptions about the variation of permeability lead to calculated moisture gradients and drying-rate curves similar to those found experimentally by many investigators.

FIG. 11. MOISTURE GRADIENTS, VARIABLE PERMEABILITY

Slab, half-thickness $4\frac{1}{2}$, $\Delta x = 1$, $s = 1$, $k = 2.5 \times 10^{-4}$, vaor pressure as in Fig. 3, permeability as in Fig. 9

FIG. 12. EFFECT OF SHAPE OF VAPOR-PRESSURE ISOTHERM ON DRYING RATE

Slab, half-thickness $4\frac{1}{2}$, $\Delta x = 1$, $s = 1$, k infinite, permeability as in Fig. 9. Curves *A*, *B*, and *C* correspond to isotherms *A*, *B*, and *C* in Fig. 13
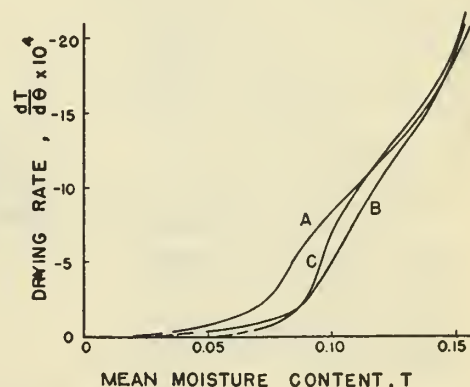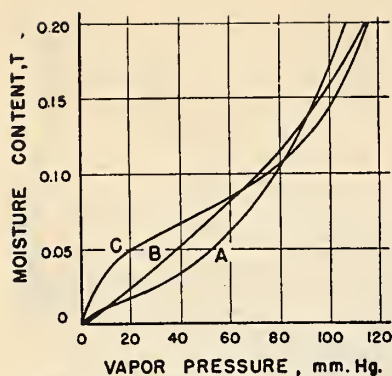
FIG. 13. VARIATIONS IN SHAPE OF VAPOR-PRESSURE ISOTHERM

Curves *A* and *B*, dehydrated carrots, curve *C*, dehydrated potatoes. Temperature, 140 degrees F.

The shapes of the drying-rate curves bear no simple relation to the quantitative variation of permeability, and no procedure for deducing the latter from the former is known.

When the permeability is variable, the effect of surface film transfer on drying rate is controlled by the ratio of the surface coefficient to the low range of permeabilities.

When the permeability is variable, the ratios of the drying rates of a slab, a long cylinder, and a sphere, of equal thickness, are no longer fixed, but depend upon the form of the permeability function.

The drying rate is inversely proportional to the square of the thickness of the piece if the surface film resistance is negligible, regardless of the form of the permeability function.

Acknowledgments

Walter Sharritts made most of the numerical computations used in this paper. Experimental drying runs
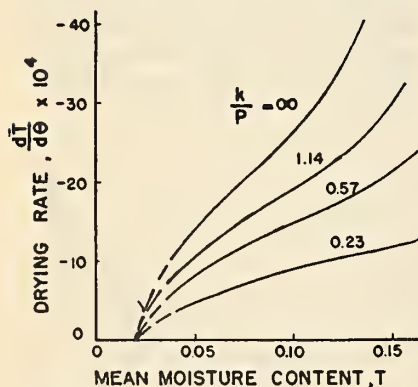

FIG. 15. EFFECT OF SURFACE FILM COEFFICIENT, PERMEABILITY VARIABLE

Slab, half-thickness 4½, $\Delta x = 1$, $s = 1$, P as in Fig. 9, vapor pressure as in Fig. 3. Solid curve, k infinite; dashed curve, $k = 2.5 \times 10^{-4}$
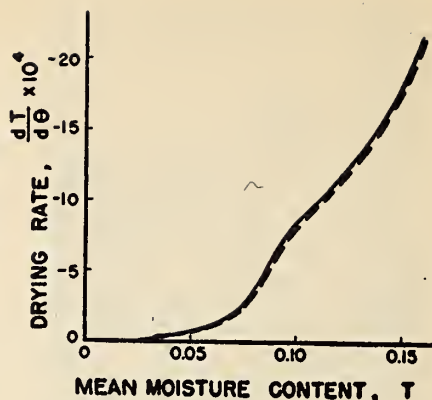

FIG. 16. EFFECT OF SHAPE OF BODY ON DRYING RATE

Vapor pressure as in Fig. 8, permeability as in Fig. 9, $s = 1$, $\Delta x = \Delta r = 1$, k infinite. Slab half-thickness and cylinder and sphere radii, 4½

FIG. 14. - EFFECT OF SURFACE FILM COEFFICIENT, PERMEABILITY CONSTANT

Slab, half-thickness 4½, $\Delta x = 1$, $s = 1$, $P = 4.4 \times 10^{-4}$, vapor pressure as in Fig. 3. Surface film coefficients, top to bottom, infinity, 5×10^{-4}, 2.5×10^{-4}, 1×10^{-4}
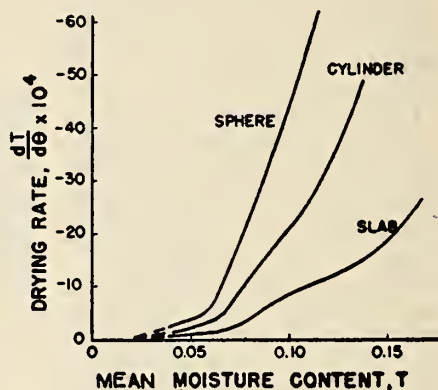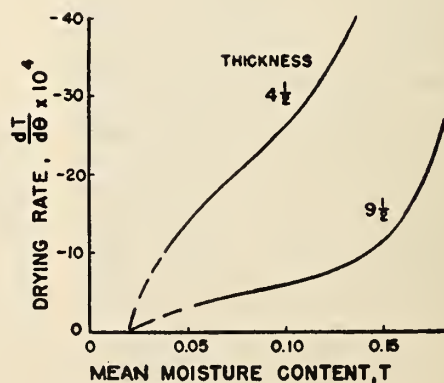
FIG. 17. EFFECT OF THICKNESS OF BODY ON DRYING RATE

Slab, $\Delta x = 1$, $s = 1$, $P = 4.4 \times 10^{-4}$, k infinite, vapor pressure as in Fig. 3
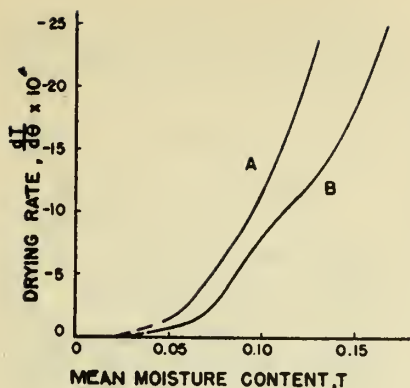
FIG. 18. EFFECT OF MIXTURE OF SIZES ON DRYING RATE

Slab, $\Delta x = 1$, $s = 1$, P as in Fig. 9, $k = 2.5 \times 10^{-4}$, vapor pressures as in Fig. 3. Curve B, half-thickness 4½. Curve A, mixture (by weight) 50 per cent standard thickness, 30 per cent ½ standard, 20 per cent ¼ standard. Rate assumed to vary as 3/2 power of thickness

In all of the calculations described in this paper a uniform initial distribution of the moisture was assumed, ($T = 0.20$). Any other desired distribution, for example parabolic, could have been used without changing the procedure or increasing the labor. The effect of the initial distribution vanishes rapidly, however, and is negligible in the drying-rate curves presented in this paper.

were made by A. H. Brown, P. W. Kilpatrick, and their coworkers. Vapor-pressure isotherms were determined by B. Makower and Fred Talburt. The author is indebted also to W. D. Ramage, Fred Stitt, H. K. Burr, and M. E. Lazar for numerous helpful suggestions.

Nomenclature

(Any consistent units)

a = ratio of inside shell radius to radial increment = $\dfrac{r_n}{\Delta r}$

C = concentration of moisture, mass per unit volume

D = diffusivity (concentration as potential)

k = surface mass transfer coefficient

p = moisture vapor pressure

p_a = partial pressure of moisture vapor in ambient air

p_n = vapor pressure at element no. n

Δp = change in vapor pressure in an increment of time

P = permeability (vapor pressure as potential)

$P_{m,n}$ = mean value of P over interval m to n

r_n = inside radius of cylindrical or spherical shell no. n

Δr = increment of radius

s = mass of dry solids per unit volume

$S_n{}^m$ = abbreviation for

$$\int_{p_n}^{p_m} P dp$$

T = moisture content, pounds of water per pound dry solids

T_c = moisture content at center line of body

T_e = moisture content in equilibrium with ambient air

T_n = moisture content at element no. n

ΔT = change in moisture content in an increment of time

Δx = increment of distance, measured normal to the drying face

$\Delta \theta$ = increment of time

$dT/d\theta$ = drying rate

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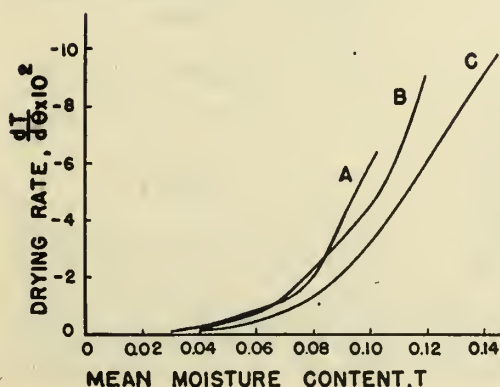22. Lewis, W. K., *Ind. Eng. Chem.* 13 (5):427-32 (1921).

FIG. 19. TYPICAL EXPERIMENTAL DRYING RATES, DEHYDRATED VEGETABLES

Curve A, Run 747, carrot half-dice, 160 degrees F., wet-bulb 90 degrees; Curve B, Run 397, 5/32-in. potato strips, 150 degrees F., wet-bulb 90 degrees; Curve C, Runs 667-9, ¼-in. potato dice, 150 degrees F., wet-bulb, 90 degrees

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Appendix

Working Equations for Approximate Calculation of Diffusion in the Square Strip and the Cube

The equations are derived by solving for ΔT the appropriate mass balances for each finite element of the body.
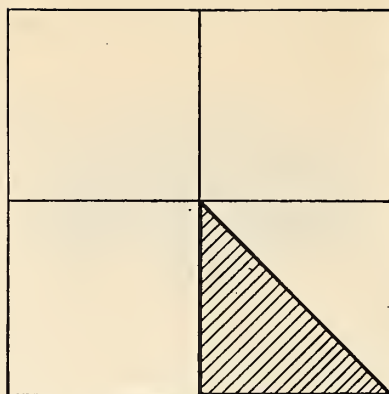

FIG. 20. TRIANGLE REPRESENTATIVE OF INFINITE SQUARE STRIP

Infinitely Long Square Strip

It is evident from considerations of symmetry that the solution is needed only for the right triangle shown in Figure 20, constituting one-eighth of the cross-section area. Figure 21 shows how the triangle will be divided into elements if the surface element has only half the thickness of the interior ones, and indicates a convenient numbering system for the elements. No diffusion will occur either across the diagonal side or across the vertical side.

In general, the mass balance for an element will contain four terms, two representing flow into the element and two flow out of it. The equation for a typical interior element, say No. 24, will be:

$$\Delta T_{24} = (S_{24}{}^{34} + S_{24}{}^{25} + S_{24}{}^{23} + S_{24}{}^{14}) \frac{\Delta\theta}{s(\Delta x)^2}$$

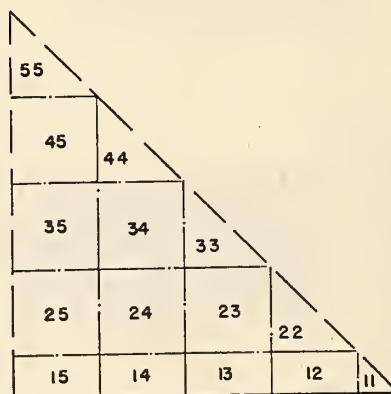The equation for a typical surface element, say No. 14, must take account of the smaller volume of the element and the smaller diffusion area from two of the adjacent elements:

FIG. 21. DIVISION OF TRIANGLE INTO ELEMENTS

$$\Delta T_{14} = 2[S_{14}{}^{24} + \tfrac{1}{2}S_{14}{}^{15} + \tfrac{1}{2}S_{14}{}^{13} + k(\Delta x)(p_a - p_{14})]\frac{\Delta\theta}{s(\Delta x)^2}$$

The remaining equations may be written in the same manner. The mean moisture content of the strip may be calculated by weighting the moisture content of each element proportionally to the volume of that element.

The Cube.

Figure 22 shows the right tetrahedron which sufficiently represents the diffusional process in the cube, and Figure 23 indicates a convenient numbering system for the elements. The tetrahedron has only 1/48 of the volume of the entire cube. The pyramidal element at the corner, Number 111, has only 1/48 of the volume of a full cubical element like Number 245, while the larger pyramidal elements along the diagonal, such as Number 333, have 1/6 of the volume of the cubical element.

In general, the mass balance for an element contains six terms, three representing inflow and three representing outflow. For an interior element, say Number 234, the equation will take the following form:

$$\Delta T_{234} = (S_{234}{}^{244} + S_{234}{}^{224} + S_{234}{}^{233} + S_{234}{}^{235} + S_{234}{}^{334} + S_{234}{}^{134}) \frac{\Delta\theta}{s(\Delta x)^2}$$

The equation for a surface element, say Number 133, taking account of the change in the element volume and of differences in diffusional area, is as follows:

$$\Delta T_{133} = 4[\tfrac{1}{2}S_{133}{}^{233} + \tfrac{1}{2}S_{133}{}^{134} + \tfrac{1}{2}S_{133}{}^{123} + \tfrac{1}{2}k(\Delta x)(p_a - p_{133})]\frac{\Delta\theta}{s(\Delta x)^2}$$

The laborious nature of the calculations for the strip and the cube is obvious. For a given fineness of subdivision there are three times as many elements to be calculated in the square strip as in the slab, and seven times
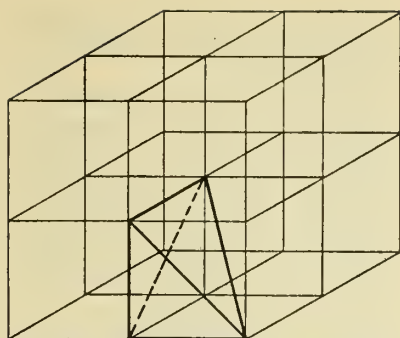
FIG. 22. TETRAHEDRON REPRESENTATIVE OF CUBE

as many in the cube as in the slab. In addition, the complexity of the calculation is greater at each element. For the fineness of subdivision shown in Figures 21 and 23 a good machine computer can complete only about twelve to fifteen steps per working day for the square strip, and only five or six for the cube. The shapes of the drying-rate curves are quite similar to those of the long cylinder and the sphere, respectively.

Presented at the Regional Meeting, San Francisco, California, August 25-28, 1946.

Discussion

G. N. Harcourt (Buffalo, N. Y.): I had the pleasure of reading Mr. Van Arsdel's well-conceived paper, and I was very much interested in it.

Unfortunately, the little I ever knew about differential equations has long since been forgotten so that part of the paper has been accepted on faith.

This kind of paper is of great potential value since it guides the thinking of others who have specific problems to solve. The specific problem may not be susceptible to solution by the mathematical processes described in this paper but the ideas that Mr. Van Arsdel, his co-workers, and other workers have set forth are of great value.

Our engineers have known for a long time that the rate of removal of moisture from material containing high percentages of moisture, 70 per cent to 10 per cent, is much greater than it is from materials containing only 10 per cent to 4 per cent moisture. For example, the condenser on a vacuum dryer must be designed to take care of the large quantity of vapor which is given off by the wet material at the beginning of the cycle.
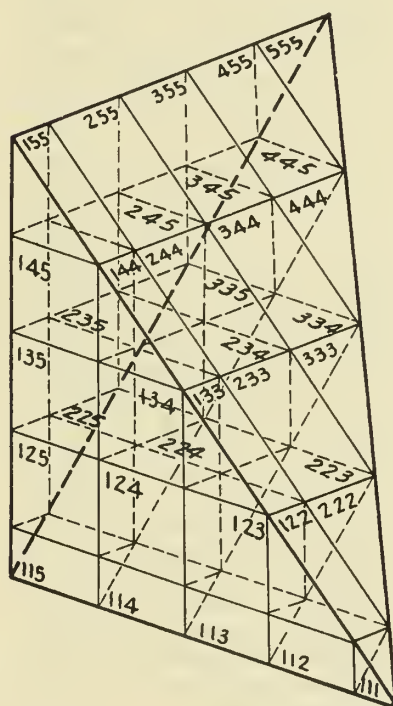

FIG. 23. DIVISION OF TETRAHEDRON INTO ELEMENTS

It is therefore our practice, when making recommendations to our customers, to keep these possibilities in mind and we will recommend on occasion the installation of two different kinds of dryers, one which will remove moisture rapidly down to the 7-10 per cent point, the other which will provide a longer retention time, the application of heat in a different way or which is in some other way more suitable for the final reduction of the moisture content, rather than to attempt to do the job on one type of dryer.

Much of our experience has been with drum dryers in which capacities are expressed in pounds of dry material per square foot of drum surface.

At one time we dried a solution of a sodium salt under the following conditions (see Table 3 also).

Moisture in solution at start = 70 per cent.

Steam pressure in drums = 60 per cent.

Our customer had specified that the dry material should contain a maximum of 0.1 per cent moisture which, as the foregoing indicates, we could not obtain economically by means of the drum dryer. We suggested finally that the drum dryer be used to reduce the moisture to 2-3 per cent at which moisture content the material from the drum dryer could be handled easily in a conveyor and did not cake. This partially dried material was then fed to a direct-fired rotary hot-air dryer where it was subjected to a very high temperature and the last traces of moisture driven off. The combination of the drum dryer and rotary dryer gave an economical and compact installation which was eminently satisfactory to the user.

TABLE 3.

Test	A	B	C	D	E	F	G	H	I	J	K
RPM drums	6-½	6-½	10	13-½	11	9	10	10	9	10¾	10
Steam pressure	44	60	60	60	60	60	60	60	60	60	60
Lb./sq.ft./hr.	4.01	4.59	5.77	9.33	6.62	5.31	5.74	5.02	5.82	6.77	6.26
Per cent moisture in dry material	2.62	0.94	1.395	10.39	2.90	1.43	1.78	1.82	2.0	4.13-5.27	2.64-3.30

Has any consideration ever been given to the effect of concentration of the soluble components in the juices in vegetables, fruits and other cellular materials while being dried. At the beginning of drying, when the material contains 70-80 per cent moisture, the solids content of the juice may be 4-6 per cent but as drying progresses the solids content will increase. It is a well-known fact that, generally, as the solids content of a solution increases, the vapor pressure of the water vapor from the solution will decrease. If the drying temperature remains constant, the rate of removal of moisture will decrease.

R. L. Huntington (University of Oklahoma, Norman): I notice a similarity between the moisture and temperature gradients for batch cooling of solids. I wonder if you made a correlation between heat flow and drying rates.

W. B. Van Arsdel: No, we have not. The special point we had in mind was the effect of a very large change in permeability and so far as I know that condition is not likely to be met in the study of heat flow. I doubt that temperature gradients in a body undergoing cooling would ever exhibit such peculiar shapes as the moisture gradients in a body undergoing drying. Conductivities and heat capacities are more nearly independent of temperature.

R. L. Huntington: You didn't make any attempt to study the effective permeability of the flow of vapor compared to that of liquid?

W. B. Van Arsdel: We begged that question. It will require carefully planned experiments to determine whether the diffusion of water under these conditions is actually proportional to a concentration difference or to a vapor-pressure difference. In an isothermal system either concept can be used, and the results will be identical. We chose to develop our results on the vapor-pressure basis, because we believe that will be more useful when we come to consider nonisothermal systems.

Written Discussion

G. M. Dusinberre (Virginia Polytechnic Institute, Blacksburg): The author points out that it is im-possible to infer from experimental drying-rate curves the details of the phenomena which combine to produce the observed result. It is possible only to test the validity of various hypotheses by comparing their logical conclusions with the experimental facts. The author has not been content to use the oversimplified classical analysis but has taken a worth-while step in testing more complex hypotheses, using the numerical analysis as a tool.

The writer has a philosophic objection to the use of the word "approximate" in the title of this paper and elsewhere, as characterizing the numerical treatment by contrast with the differential treatment. Engineers and scientists use differential equations so much, and to such great advantage, that there is a tendency to join the mathematicians in regarding these equations as fundamental, rather than the physical situation which the equations are set up to represent.

The present physical problem is to predict the effects, observable on the large scale, of discrete molecules moving in a heterogeneous and discontinuous region. As one approximation we pass over the discontinuities, treat the region as homogeneous, and apply the rules of calculus. As another approximation we set up arbitrarily discontinuities at convenience and apply the rules of arithmetic. Both methods are approximate, physically. Both are exact, mathematically, in the sense that there is a demonstrable relation between the initial assumptions and the final solution. But the assumptions are slightly different in the two cases. It is only when the one solution is compared with the other set of assumptions that it deserves to be called an approximation.

No one, least of all the writer, can overlook the great advantage of an analytical solution, when one is obtainable. This field of mathematics needs no apology of defense. Neither does it need the undeserved halo of being called "exact" in its physical application. The numerical treatment evidently, at this time, needs some defense and explanation. But it does not deserve to be characterized as merely approximate, by contrast with the other.

For the engineer and scientist the test of accuracy is agreement with experiment. From this viewpoint, the author has done some excellent work in demonstrating the accuracy of the method which he refers to still as approximate.

Incidentally, it may be borne in mind that an analytical solution is usually in the form of an infinite series. If the first few steps of a numerical solution are inaccurate, so are the first few terms of a series. If the numerical solution may run into a divergence, so may a series. But there are always remedies.

The author stated that the writer's methods were unsatisfactory when variation of diffusivity was wide and abrupt. This offers difficulty with any treatment, but it may be pointed out that the $\int P d p$ used by the author is exactly analogous to the $\int k d T$ used by the writer. Thus, whatever advantages or disadvantages are found must lie in the details of application, since the two treatments are the same in principle.

W. B. Van Arsdel: Professor Dusinberre's point about my designation of the numerical method as "approximate" is very well taken. The approximations with which we should be seriously concerned are those concealed within the simplifying assumptions. I suspect that many of us are unduly impressed by the "elegance" of analytical solutions. Numerical solutions look, in contrast, clumsy and unimaginative; unconsciously, then, we assume they must be less precise representations of the true physical situation.

The comment made in the paper about Professor Dusinberre's semigraphical method for handling variable-diffusivity calculations was perhaps not sufficiently explained. The principle is, as he says, exactly the same as the one I have applied. However, if a radical change in diffusivity or permeability is involved (such as the 20- to 40-fold changes discussed in the paper) the construction of his "correction chart" to a sufficient degree of precision becomes exceedingly laborious in itself. More pertinent from the standpoint of the exploratory work reported here is the fact that many different combinations of permeability function and vapor-pressure isotherm were being investigated. Each different combination would have required a new correction chart. If a large number of calculations were to be made for a single substance the labor invested in preparation of the chart might be justified.